

## REMARKS

Applicant has corrected pages 1, 2 and 3 of the specification as suggested by the Examiner. Accordingly, the objection to the disclosure relative to pages 1, 2 and 3 should be withdrawn.

Applicants have submitted an executed Declaration signed by each of the inventors to clarify the objections raised both to the specification and claims. There is no inconsistency in the reported afterglow time between the table and the examples in the specification. The intent is to show that the light emitting material of the present invention has excellent visible afterglow time. More particularly, as pointed out in paragraph 6 of the Declaration, the light emitting material showed a brightness of about 8500 mcd/m<sup>2</sup> after five seconds at which time the irradiation was stopped. The visible afterglow time of the samples were then measured in a conventional manner. Table 1 shows a brightness at the 480<sup>th</sup> minute after stopping irradiation of more than 10 mcd/m<sup>2</sup>. The number of hours for the brightness to further decay down to 3 mcd/m<sup>2</sup> for each of the examples totaled above 80 hours as set forth on page 7 of the specification. Hopefully, this should explain why the table and examples are in fact consistent with each other.

Paragraph 4 of the Declaration addresses the question raised by the Examiner regarding the lack of clarity between the two embodiments of the examples having the same formulas when each start with different amounts of raw material. Contrary to the allegation of the Examiner, the formulas for the two examples shown in the present application are not identical. The value of X is identical in each of the examples ( $X = 0.01$ ) but the number X does not determine the relative individual amounts of Sr, Eu and Dy. Instead, the fixed X value determines only the total amount of all three elements when in combination. The individual amounts of the elements may still vary relative to one another and in fact are changed even though the total amounts of all three elements Sr, Eu and Dy in combination remain the same. This results in a difference between the first and second examples which resides in the different individual

amounts of SrCO<sub>3</sub>, EU<sub>2</sub>O<sub>3</sub> and Dy<sub>2</sub>O<sub>3</sub>. Paragraph 4 of the executed Declaration should resolve the lack of clarity that the Examiner may have believed existed in the two embodiments of the examples. Accordingly, this objection should now be withdrawn.

Paragraph 2 of the executed Declaration addresses the question raised by the Examiner concerning the teaching in the specification of the ratio of the two phases and Paragraph 3 explains why the ratio of the A-phase to the B-phase in the resultant is 1:1. This ratio is determined by X-ray diffraction which is a measurement of the light-emitting spectrum of both phases respectively. No coefficient is shown in the formula and the compound as claimed consists of an inseparable mixture of each phase in a ratio of 1 to 1. Accordingly, the suggested revision by the Examiner is incorrect and based on a misinterpretation of the invention.

As regards the use of the dot in the formula separating the two phases, it is considered necessary for distinguishing a complex bi-phase crystalline compound from a single compound as is taught in the reference cited by the Examiner. Moreover, applicants – as experts in the art and as the inventors – are certainly entitled to define a formula representing the complex compound as they understand it and not to be relegated to how the Examiner interprets it. Applicant has not misused the symbol in the formula and has clearly explained its significance. Moreover, as set forth by applicants in the executed Declaration, the use of a dot “.” in the formula separating the two phases of the material is commonly used in crystallography when the compound is a complex compound. Accordingly, the objection to the use of the dot in the formula should be withdrawn.

Applicant has amended claims 12 and 13 as suggested by the Examiner to overcome the objection based upon the existence of certain informalities. Accordingly, the objection to claims 12 and 13 based upon such informalities should now be withdrawn.

The rejection of claims 11, 12 and 16 under 35 USC 112, first paragraph, as containing subject matter which was not described in the specification in such a way as to reasonably convey to one skilled in the relevant art that the inventors at the time the application was filed had possession of the claimed invention is respectfully traversed. Much of this rejection is based upon the misunderstanding of the Examiner which applicant believes has been resolved by the filing of the executed Declaration. Moreover, claim 11 has been amended to specify that the element B exists in the entire crystallization structure of both phases consistent with page 4 of the specification.

Claim 12 has been amended in conformity with page 3 of the specification and requires the mixture to be heated from 850°C to 1200°C for three hours under a reduction condition and thereafter for the temperature of the heated mixture to be maintained relatively constant to form a sintered body. The rejection of claim 12 under 35 USC 112, first paragraph, should now be withdrawn. Claim 16 is dependent upon claim 12 and is consistent with the specification. Accordingly, the rejection of claim 16 under 35 USC 112, first paragraph should also be withdrawn.

The rejection of claims 12-16 under 35 USC 112, first paragraph, based on lack of enablement is respectfully traversed. Applicant believes that the amendment to claim 12 overcomes this rejection in that the temperature of the heated mixture is being maintained relatively constant so as to form a sintered body. Forming the sintered body is a well known procedure to those skilled in the art. Accordingly, the specification clearly enables any person skilled in the art following the method of claims 12 to 16 to make the invention. This rejection should now be withdrawn.

The rejection of claims 10-12 under 35 USC 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicants regard as the invention is respectfully traversed.

The executed Declaration attached to this amendment addresses in paragraph 4 the issue of indefiniteness raised by the Examiner and was explained

heretofore. As is explained in paragraph 3 of the Declaration, the ratio of the A-phase to the B-phase in the resultant is 1:1 i.e. the compound consists of an inseparable mixture of each phase in a 1 to 1 ratio. Accordingly, and from the explanation in applicants' Declaration, claims 10 and 11 are both clear and definite. This rejection should therefore be withdrawn.

Claim 10 has been amended to remove the indefinite expression "at substantially 1200°C". Accordingly, the rejection of this claim under 35 USC 112 should be withdrawn.

The rejection of claims 10 and 11 under 35 USC 112, second paragraph as failing to set forth the subject matter which applicants' regard as their invention is respectfully traversed. Once again, this is based upon a misunderstanding of the invention. Claim 10 defines a light-emitting material having a paragenesis crystalline structure consisting of two different phases expressed by a particular formula with the elements Sr, Eu and Dy identified by weight. The formula as explained by the applicants in the attached Declaration defines a complex bi-phase crystalline compound consisting of an A-phase having the formula  $(\text{Sr},\text{Eu},\text{Dy})_{0.95+\underline{x}}(\text{A1},\text{B})_2\text{O}_{3.95+\underline{x}}$  and a B-phase having the formula  $(\text{Sr},\text{Eu},\text{Dy})_{4.\underline{x}}(\text{A1},\text{B})_{14}\text{O}_{25.\underline{x}}$ . This is consistent with the specification. Accordingly, the rejection of claims 10 and 11 under 35 USC 112, second paragraph, is without merit and should be withdrawn.

The rejection of claims 12 and 13 under 35 USC 103(a) as being unpatentable over Royce, et al is respectfully traversed.

Royce, et al does not teach a pulverized mixture of the same raw materials and accordingly, no basis exists to support the allegation that it is a light-emitting material consisting of a different combination of pulverized raw materials. The cases cited by the Examiner relate to processes in which the compositions are identical except for the numerical range of the elements. In claim 12, the raw materials consist of  $\text{SrCO}_3$ ,  $\text{H}_3\text{BO}_3$ ,  $\text{Eu}_2\text{O}_3$  and  $\text{Dy}_2\text{O}_3$  whereas Royce, et al '303 does not teach a single mixture

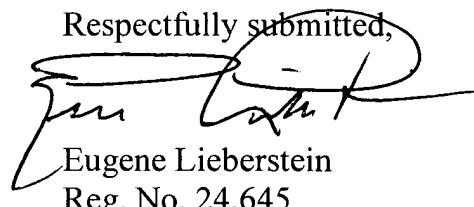
consisting of these elements. Accordingly, the rejection of claims 12 and 13 under 35 USC 103(a) should be withdrawn.

The rejection of claims 12-15 under 35 USC 103(a) as being obvious over Hao, et al is respectfully traversed. As set forth in paragraph 1 of the attached Declaration, the inventors and assignees of the cited Hao, et al reference, U.S. Patent 5,885,483, are identical to the inventors and assignees of the present U.S. patent application, serial number 09/646,978. Moreover, the subject application was filed after November 29, 1999 in accordance with MPEP 706.02 (I)(1) and 7606.02(I)(2).

Accordingly, the rejection of claims 12-15 as being obvious over Hao, et al should be withdrawn.

Reconsideration and allowance of claims 10-16 is respectfully solicited.

Respectfully submitted,



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#### MAILING CERTIFICATE

I hereby certify that this correspondence is being deposited with the U.S. Postal Service as first class mail in an envelope addressed: Commissioner of Patents & Trademarks, Washington, DC 20231 on April 23, 2003.



Date: 4/23/03

## LIGHT-EMITTING MATERIAL AND PRODUCING METHOD THEREOF

### TECHNICAL FIELD

The present invention relates to a light-emitting material and a producing method thereof, and more particularly, to an inorganic light-emitting material using a rare-earth element as an exciting agent and having an afterglow time and a producing method thereof.

### BACKGROUND TECHNIQUE

A light-emitting material is utilized is mixed in ink or paint to make light emitting paint, and is utilized on a safe sign and a clock board. Conventionally, copper-excited zinc sulfide (ZnS:Cu) was widely used as the light-emitting material. ZnS:Cu has high light-emitting efficiency in the light-emitting spectral region, but its brightness is lowered extremely rapidly, and the visible afterglow time is as short as 20 to 30 minutes. When the ZnS:Cu is exposed to ultraviolet rays in areas exposed to moisture, decomposition and degeneration are generated and the body color of the material darkens. Therefore, there are constraints for using ZnS:Cu outside, and optimal material as a substitute for ZnS:Cu had long been required.

In CN1053807A, a light-emitting material ( $m(\text{Sr}_{1-x}\text{Eu}_x)\text{O}\cdot n\text{Al}_2\text{O}_3\cdot y\text{B}_2\text{O}_3$ ) having long afterglow ability and is laid open. In the above general formula,  $1 \leq m \leq 5$ ,  $1 \leq n \leq 8$ ,  $0.005 \leq y \leq 0.35$  and  $0.001 \leq x \leq 0.1$ . The afterglow time of this light-emitting material is in a range of from 10 to 20 hours.

In [USP5, 376,303,] USP 5,376,303, phosphor having long afterglow ability comprises a compound  $(\text{MO}\cdot a(\text{Al}_{1-b}\text{B}_b)_2\text{O}_3\cdot c\text{R})$ .

In this general formula,  $0.5 \leq a \leq 10.0$ ,  $0.0001 \leq b \leq 0.5$ ,  $0.0001 \leq c \leq 0.2$ , MO is at least one compound selected from a group consisting of MgO, CaO, SrO and ZuO. R consists of Eu and at least one additive rare-earth element selected from a

group consisting of Pr, Nd, RE and Tm.

In the above-mentioned patent, some phosphors such as  $[SrO \cdot 2.10 (Al_{10.952}B_{0.048})_2O_3 : 0.005Eu, 0.020Dy]$   $SrO \cdot 2.10 (Al_{10.952}B_{0.048})_2O_3 : 0.005Eu, 0.020Dy$  (which will be referred to as "A" hereinafter), and  $[SrO \cdot 1.025 (Al_{0.976}B_{0.024})_2O_3 : 0.005Eu, 0.015Dy]$   $SrO \cdot 1.025 (Al_{0.976}B_{0.024})_2O_3 : 0.005Eu, 0.015Dy$  (which will be referred to as "B" hereinafter) were prepared, and residual light-emitting time and brightness of these phosphors were evaluated in view of afterglow time constant (n) and relative brightness (ZnS: Cu, Cl were evaluated). A measuring result shows that the afterglow time constants of A and B phosphors and ZnS: Cu, Cl are 0.94, 0.86 and 1.26, respectively, and phosphorus brightness after 10 seconds to be 144, 220 and 100, respectively, and phosphorus brightness after 20 seconds was 934, 1320 and 100, respectively.

It is found that these materials are clearly improved as compared with ZnS: Cu, Cl, but they are not yet in practical use.

Thereupon, the present inventors conducted various researches for producing light-emitting material using rare-earth element Eu as the light-emitting material. Based on the researches, the present inventors found that a light-emitting material having a new crystallization structure was obtained by adding an appropriate amount of B and an additive exciting agent. This light-emitting material has a desired long afterglow time and high brightness.

Therefore, a first objective of the present invention is to provide a light-emitting material having a long afterglow time and high brightness.

Moreover, a second object of the present invention is to provide a producing method of the above-mentioned light-emitting material.

## DISCLOSURE OF THE INVENTION

To achieve the objects, the present invention provides the following light-emitting material and the producing method.

That is, a light-emitting material of the present invention includes a dipophase compound crystallization expressed in a general formula:  $(\text{Sr}, \text{Eu}, \text{Dy})_{0.95 \pm x}(\text{Al}, \text{B})_2\text{O}_{3.95 \pm x}$   $(\text{Sr}, \text{Eu}, \text{Dy})_{4-x}(\text{Al}, \text{B})_{14}\text{O}_{25-x}$  ( $x=0.01 \sim 0.1$ ).

This dipophase compound has a new crystallization structure and consists of two phases, i.e.,  $(\text{Sr}, \text{Eu}, \text{Dy})_{0.95 \pm x}(\text{Al}, \text{B})_2\text{O}_{3.95 \pm x}$  and  $(\text{Sr}, \text{Eu}, \text{Dy})_{4-x}(\text{Al}, \text{B})_{14}\text{O}_{25-x}$ . This conclusion was obtained from XRD (X-ray diffraction) analysis using a Chinese geological university material scientific divisional crystallization structure and a large amount of samples carried out in a crystallization chemical laboratory. Based on the XRD analysis, and using an optical microscope and electronic probe analytical technique, it was corroborated that the above two phases are symbiosis and have light-emitting function.

The producing method of producing a light-emitting material includes the following steps:

- (1) step for measuring previously pulverized raw materials, and mixing them to obtain a mixture of raw material,
- (2) step for putting the mixture into a container, heating the mixture from 850°C to 1200°C for three hours under a reduction condition, keeping the temperature for five to six hours, thereby obtaining a sintered body,
- (3) step for stopping the heating operation and cooling the sintered body nature down to room temperature, and
- (4) step for pulverizing the sintered body to obtain a product.

According to the light-emitting material and the producing method of the present invention, there is a light-emitting effect that which visible long afterglow ability as compared with a similar light-emitting material.

## **BEST MODE FOR CARRYING OUT THE INVENTION**

In an assessment process concerning a phase of a light-emitting material of the present invention, using analytical means such as an X-ray fluorescent analysis, a plasma spectral analysis, electronic probe and X-ray photoelectron

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In this general formula,  $0.5 \leq a \leq 10.0$ ,  $0.0001 \leq b \leq 0.5$ ,  $0.0001 \leq c \leq 0.2$ ,  $MO$  is at least one compound selected from a group consisting of  $MgO$ ,  $CaO$ ,  $SrO$  and  $ZnO$ .  $R$  consists of Eu and at least one additive rare-earth element selected from a

group consisting of Pr, Nd, ~~Y~~ and Tm.

In the above-mentioned patent, some phosphors such as  $\text{SrO} \cdot 2.10 (\text{Al}_{10.952}\text{B}_{0.048})_2\text{O}_3$ : 0.005Eu, 0.020Dy (which will be referred to as "A" hereinafter), and  $\text{SrO} \cdot 1.025 (\text{Al}_{0.976}\text{B}_{0.024})_2\text{O}_3$ : 0.005Eu, 0.015Dy (which will be referred to as "B" hereinafter) were prepared, and residual light-emitting time and brightness of these phosphors were evaluated in view of afterglow time constant ( $n$ ) and relative brightness (ZnS: Cu, Cl were evaluated). A measuring result shows that the afterglow time constants of A and B phosphors and ZnS: Cu, Cl are 0.94, 0.86 and 1.26, respectively, and phosphorus brightness after 10 seconds to be 144, 220 and 100, respectively, and phosphorus brightness after 20 seconds was 934, 1320 and 100, respectively.

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- (3) step for stopping the heating operation and cooling the sintered body nature down to room temperature, and
- (4) step for pulverizing the sintered body to obtain a product.

According to the light-emitting material and the producing method of the present invention, there is a light-emitting effect that which visible long afterglow ability as compared with a similar light-emitting material.

## **BEST MODE FOR CARRYING OUT THE INVENTION**

In an assessment process concerning a phase of a light-emitting material of the present invention, using analytical means such as an X-ray fluorescent analysis, a plasma spectral analysis, electronic probe and X-ray photoelectron

## AMENDMENTS TO THE CLAIMS

**11. (Amended)** A light-emitting material according to claim 10, wherein the element B exists [entirely in crystalline form in both of the two] in the entire crystallization structure of both phases.

**12. (Amended)** A method of producing a light-emitting material comprising the steps of:

- (1) pulverizing raw material(s) from the group consisting of [ $\text{SrCO}_3$ ,  $\text{AO}_2\text{O}_3$ ,  $\text{H}_3\text{BO}_3$ ,  $\text{Eu}_2\text{O}_3$  and  $\text{Dy}_2\text{O}_3$ ]  $\text{SrCO}_3$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{H}_3\text{BO}_3$ ,  $\text{Eu}_2\text{O}_3$  and  $\text{Dy}_2\text{O}_3$ ,
- (2) heating the pulverized raw material(s) at a temperature in the range between 850°C and 1200°C for three hours under a reduction condition,
- (3) maintaining the temperature of step 2 relatively constant [at substantially 1200°C] to form a sintered body,
- (4) cooling the sintered body down to room temperature, and
- (5) pulverizing the sintered body.

**13. (Amended)** The method of claim 12 wherein [ $\text{Eu}_3^+$ ]  $\text{Eu}^{3+}$  of  $\text{Eu}_2\text{O}_3$  is reduced to [ $\text{Eu}_2^+$ ]  $\text{Eu}^{2+}$  during sintering.

**AMENDED VERSION OF THE CLAIMS**

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**11.** A light-emitting material according to claim 10, wherein the element B exists in the entire crystallization structure of both phases.

**12.** A method of producing a light-emitting material comprising the steps of:

- (1) pulverizing raw material(s) from the group consisting of SrCO<sub>3</sub>, Al<sub>2</sub>O<sub>3</sub>, H<sub>3</sub>BO<sub>3</sub>, Eu<sub>2</sub>O<sub>3</sub> and Dy<sub>2</sub>O<sub>3</sub>;
- (2) heating the pulverized raw material(s) at a temperature in the range between 850°C and 1200°C for three hours under a reduction condition,
- (3) maintaining the temperature of step 2 relatively constant to form a sintered body,
- (4) cooling the sintered body down to room temperature, and
- (5) pulverizing the sintered body.

**13.** The method of claim 12 wherein Eu3+ of Eu<sub>2</sub>O<sub>3</sub> is reduced to Eu2+ during sintering.

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